

We claim:

1. A process for heterogeneously catalyzed partial direct oxidation of propane and/or isobutane to at least one of the target products acrylic acid, methacrylic acid, by feeding a starting reaction gas mixture comprising propane and/or isobutane, molecular oxygen and at least one inert diluent gas and having a inlet pressure P^1 to a reaction stage which, apart from an inlet for the starting reaction gas mixture, optionally further inlets for auxiliary gases, and an outlet for the product gas mixture, is sealed on the gas side, in the reaction stage directly oxidizing the propane and/or isobutane present in the starting reaction gas mixture partially to at least one target product by passing the starting reaction gas mixture at elevated temperature over a solid state catalyst, and conducting the reaction gas mixture as a product gas mixture comprising at least one target product and having the outlet pressure P^2 out of the reaction stage and, with this pressure P^2 , into a workup stage which, apart from an inlet for the product gas mixture, optionally further inlets for auxiliary gases, and an outlet for the residual product gas mixture, is sealed on the gas side, in the workup stage basically separating target product present in the product gas mixture of the reaction stage from said product gas mixture into a liquid phase and conducting the remaining residual product gas mixture which comprises not only propane and/or isobutane and also in some cases propene and/or isobutene and has the outlet pressure P^3 , where $P^3 < P^1$, out of the workup stage and recycling propane and/or isobutane present in the residual product gas mixture into the reaction stage, which comprises selecting P^1 in such a way that $P^3 \geq 1.5$ bar and dividing the residual product gas mixture into two portions of the same composition and discharging one portion as output and recycling the other portion as cycle gas and feeding it back to the reaction stage, compressed to the inlet pressure P^1 , as a constituent of the starting reaction gas mixture.
2. A process as claimed in claim 1, wherein the residual product gas mixture contains at least 5% by volume of constituents other than propane and/or isobutane and also other than propene and/or isobutene.
3. A process as claimed in claim 1, wherein the residual product gas mixture contains at least 10% by volume of constituents other than propane and/or isobutane and also other than propene and/or isobutene.
4. A process as claimed in any of claims 1 to 3, wherein the pressure $P^3 \geq 1.5$ bar and ≤ 25 bar.

5. A process as claimed in any of claims 1 to 3, wherein the pressure $P^3 \geq 1.5$ bar and ≤ 20 bar.
6. A process as claimed in any of claims 1 to 3, wherein the pressure $P^3 \geq 1.5$ bar and ≤ 10 bar.
- 5 7. A process as claimed in any of claims 1 to 3, wherein the pressure $P^3 \geq 2$ bar and ≤ 8 bar.
- 10 8. A process as claimed in any of claims 1 to 7, wherein the pressure P^1 is from 1 to 4 bar above the pressure P^3 .
9. A process as claimed in any of claims 1 to 7, wherein the pressure P^1 is from 1.5 to 3.5 bar above the pressure P^3 .
- 15 10. A process as claimed in any of claims 1 to 9, wherein P^1 is from 3 to 10 bar.
11. A process as claimed in any of claims 1 to 9, wherein P^1 is from 4 to 8 bar.
- 20 12. A process as claimed in any of claims 1 to 11, wherein the portion of the residual product gas mixture which is discharged as output is discharged via an expander.
13. A process as claimed in any of claims 1 to 12, wherein the reaction stage is a catalyst-charged tube bundle reactor or fluidized bed reactor.
- 25 14. A process as claimed in any of claims 1 to 13, wherein the workup stage is an absorption column or a column for fractional condensation or a series arrangement of quench stages.
- 30 15. A process as claimed in any of claims 1 to 14, wherein the active composition of the catalyst is a mult-metal oxide composition which comprises the elements Mo, V, at least one of the two elements Te and Sb, and at least one of the elements from the group consisting of Nb, Ta, W, Ti, Al, Zr, Cr, Mn, Ga, Fe, Ru, Co, Cs, Ca, Sr, Ba, Rh, Ni, Pd, Pt, La, Pb, Cu, Re, Ir, Y, Pr, Nd, Tb, Bi, B, Ce, Sn, Zn, Si, Na, Li, K, Mg, Ag, Au and In in combination.
- 35 16. A process as claimed in any of claims 1 to 14, wherein the active composition of the catalyst is a mult-metal oxide composition which contains the element combination having the stoichiometry I

where

- 5 M¹ = Te and/or Sb,
M² = at least one of the elements from the group consisting of Nb, Ta, W, Ti,
Al, Zr, Cs, Ca, Sr, Ba, Cr, Mn, Ga, Fe, Ru, Co, Rh, Ni, Pd, Pt, La, Bi, Pb,
Cu, Re, Ir, Y, Pr, Nd, Tb, Ce, Sn, Zn, Si, Na, Li, K, Mg, Ag, Au and In,
10 b = from 0.01 to 1
c = from > 0 to 1 and
d = from > 0 to 1.

15 17. A process as claimed in any of claims 1 to 16, wherein the oxygen source used is air.

18. A process as claimed in any of claims 1 to 17, wherein the reaction temperature is from 200 to 700°C.

20 19. A process as claimed in any of claims 1 to 18, wherein the starting reaction gas mixture contains

from 0.5 to 15% by volume of propane or isobutane,
from 10 to 90% by volume of air,
from 0 to 50% by volume of steam and
a remainder of cycle gas.

25 20. A process as claimed in any of claims 1 to 18, wherein the starting reaction gas mixture contains

from 0.5 to 15% by volume of propane or isobutane,
from 10 to 90% by volume of air,
from 10 to 50% by volume of steam and
a remainder of cycle gas.

30 30. A process as claimed in any of claims 1 to 18, wherein the starting reaction gas mixture contains

from 0.5 to 15% by volume of propane or isobutane,
from 10 to 90% by volume of air,
from 10 to 50% by volume of steam and
a remainder of cycle gas.

35 35. 21. A process as claimed in any of claims 1 to 18, wherein the starting reaction gas mixture contains

from 70 to 90% by volume of propane or isobutane,
from 5 to 25% by volume of molecular oxygen,
from 0 to 25% by volume of steam and

a remainder of cycle gas.

22. A process as claimed in any of claims 1 to 21, wherein the conversion from propane and/or isobutane, based on single pass of the reaction gas mixture through the reaction stage, is from 10 to 70 mol%.
- 5 23. A process as claimed in claim 22, wherein the selectivity of the target product formation is from 40 to 98 mol%.
- 10 24. A process as claimed in any of claims 1 to 23, wherein the target product present in the product gas mixture of the reaction stage is basically separated into the liquid phase in such a way that the molar ratio W of the steam present in the remaining residual product gas mixture to the propane present therein is at least 50% smaller than the corresponding molar ratio W' in the product gas mixture of the reaction stage.
- 15 25. The process as claimed in any of claims 1 to 24, wherein the target product present in the product gas mixture of the reaction stage is basically separated into the liquid phase in an absorption column by absorption into an organic solvent in such a way that the discharge from the absorption column is monophasic.
- 20 26. A process as claimed in any of claims 1 to 25, wherein the propane and/or isobutane and also any propene and/or isobutene present in the portion of the residual product gas mixture which is discharged as output are removed from said residual product gas mixture and recycled into the reaction stage, recompressed to the inlet pressure P¹.
- 25 27. A process as claimed in any of claims 1 to 26, wherein the ratio V of that portion of the residual product gas mixture which is recycled as cycle gas to that portion of the residual product gas mixture which is discharged as output is ≥ 0.5 and ≤ 30.
- 30 28. A process as claimed in any of claims 1 to 26, wherein the ratio V of that portion of the residual product gas mixture which is recycled as cycle gas to that portion of the residual product gas mixture which is discharged as output is ≥ 2 and ≤ 25.
- 35 29. A process as claimed in any of claims 1 to 26, wherein the ratio V of that portion of the residual product gas mixture which is recycled as cycle gas to that portion of the residual product gas mixture which is discharged as output is ≥ 3 and ≤ 20.

30. A process as claimed in any of claims 1 to 29, wherein the cycle gas is recompressed to the inlet pressure P^1 using a blower.
- 5 31. A process as claimed in any of claims 1 to 30, wherein the oxygen source used is air which is compressed to the inlet pressure P^1 by means of a radial compressor.
32. A process as claimed in any of claims 1 to 31, which is a process for partial direct oxidation of propane to acrylic acid.